Linear Viscoelastic and Dielectric Relaxation Response of Unentangled UPy-Based Supramolecular Networks - DTU Orbit (09/11/2017)

Linear Viscoelastic and Dielectric Relaxation Response of Unentangled UPy-Based Supramolecular Networks

Supramolecular polymers possess versatile mechanical properties and a unique ability to respond to external stimuli. Understanding the rich dynamics of such associative polymers is essential for tailoring user-defined properties in many products. Linear copolymers of 2-methoxyethyl acrylate (MEA) and varying amounts of 2-ureido-4[1*H*]-pyrimidone (UPy) quadruple hydrogen-bonding side units were synthesized via free radical polymerization. Their linear viscoelastic response was studied via small amplitude oscillatory shear (SAOS). The measured linear viscoelastic envelope (LVE) resembles that of a well-entangled polymer melt with a distinct plateau modulus. Dielectric relaxation spectroscopy (DRS) was employed to independently examine the lifetime of hydrogen bond units. DRS reveals a high frequency α-relaxation associated with the dynamic glass transition, followed by a slower α*-relaxation attributed to the reversible UPy hydrogen bonds. This time scale is referred to as the bare lifetime of hydrogen bonding units. Using the sticky Rouse model and a renormalized lifetime, we predict satisfactorily the LVE response for varying amounts of UPy side groups. The deviation from the sticky Rouse prediction is attributed to polydispersity in the distribution of UPy groups along the chain backbone. We conclude that the response of associating polymers in linear viscoelasticity is general and does not depend on the chemistry of association, but rather on the polymer molecular weight (MW) and MW distribution, the number of stickers per chain, n_S, and the distribution of stickers along the backbone.

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